

Direct Electrochemical Conversion of Carbon Anode Fuels in Molten Salt Media

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DIRECT ELECTROCHEMICAL CONVERSION OF CARBON ANODE FUELS IN MOLTEN SALT MEDIA

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We are conducting research into the direct electrochemical conversion of reactive carbons into electricity—with experimental evidence of total efficiencies exceeding 80% of the heat of combustion of carbon. Together with technologies for extraction of reactive carbons from broad based fossil fuels, direct carbon conversion addresses the objectives of DOE's "21st Century Fuel Cell" with exceptionally high efficiency (>70% based on standard heat of reaction, ΔH_{std}), as well as broader objectives of managing CO₂ emissions. We are exploring the reactivity of a wide range of carbons derived from diverse sources, including pyrolyzed hydrocarbons, petroleum cokes, purified coals and biochars, and relating their electrochemical reactivity to nano/microstructural characteristics.

We are developing and optimizing a cell in which carbon fuels are directly converted electrochemically into electrical energy, as shown conceptually in Figure 1. The rate of conversion of the carbons (mA/cm^2) indicates their relative reactivity. Our electrochemical cell uses a $\text{Li}_2\text{CO}_3/\text{K}_2\text{CO}_3$ (46.6/53.4 wt %, 38/62 mole %) carbonate electrolyte and oxygen (air) in the cathode compartment, and a slurry consisting of molten carbonate electrolyte and carbon fuel in the anode compartment. A porous ceramic separator transports dissolved CO_2 and carbonate ions between the two compartments allowing the cathodic half reaction: $\text{O}_2 + 2\text{CO}_2 + 4\text{e}^- = 2\text{CO}_3^{2-}$ and the anodic half reaction: $\text{C} + 2\text{CO}_3^{2-} = 3\text{CO}_2 + 4\text{e}^-$ to occur. The net reaction is $\text{C} + \text{O}_2 = \text{CO}_2$.

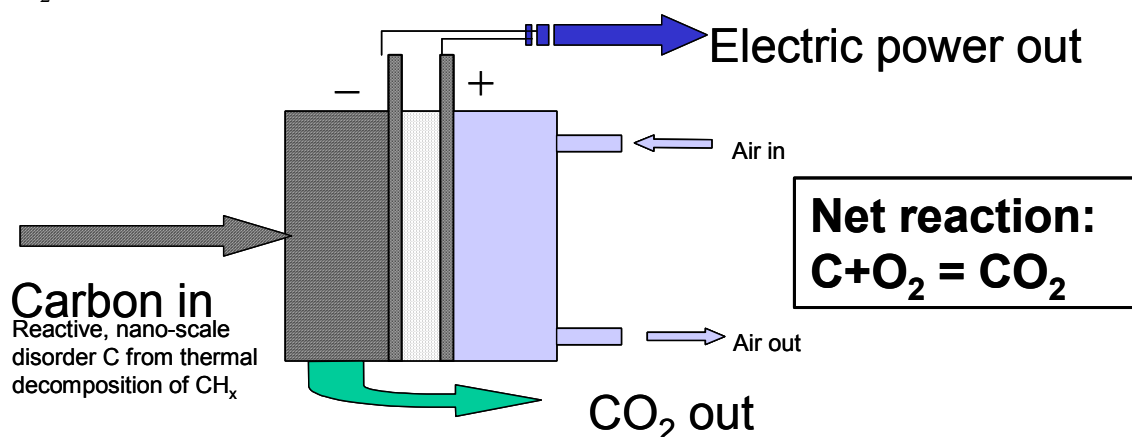


Figure 1. The carbon direct conversion cell uses submicron particles of carbon distributed pneumatically to cells, a molten carbonate electrolyte, and conventional oxygen reduction cathodes. The pure CO_2 byproduct may be sequestered without further purification or separation, or reused in enhanced oil or gas recovery.

We have demonstrated working rates as high as $100 \text{ mA}/\text{cm}^2$ at 0.8 V and $200 \text{ mA}/\text{cm}^2$ at 0.5 V with a particularly reactive fuel, Carbon 1. Figure 2 shows dramatically different power density curves for three carbon samples, Carbons 1, 2 and 3. The more power per area in the same electrochemical cell, the more reactive the carbon. We gain important insight into the basis for electrochemical reactivity by correlating structural characteristics of the carbon samples with their reactivities.

We have characterized candidate fuels using several techniques: (1) Transmission electron microscopy reveals primary particle size and aggregate size, (2) BET (N_2) adsorption provides specific surface area measurements, (3) X-ray diffraction exposes the layer spacing (d -spacing) and relative lattice disorder (L_c), (4) Thermal gravimetric analysis (TGA) shows chemical reactivity to air oxidation. Small L_c values and low decomposition temperatures and high surface area are correlated with high reactivity. Some of these measurements for Carbons 1, 2 and 3 are shown in Table I.

Future work includes engineering and assembly of a scaled-up direct carbon conversion cell, capable of producing 0.1 kW per (stackable) cell. We will also continue to study carbon fuels of different origin and characteristics for reactivity and structure.

Table I. Structural characteristics of carbon samples correlate with their electrochemical reactivity.

Sample	d(002) spacing (nm)	L_c (nm)	Surface Area (m^2/g)	Temp. at 80% Decomp. ($^{\circ}\text{C}$)	(mA/cm^2) at 0.8 V
Carbon 3	0.369	1.7	60	731	103
Carbon 2	0.374	2.9	75	819	30
Carbon 1	0.363	4.3	9	796	6

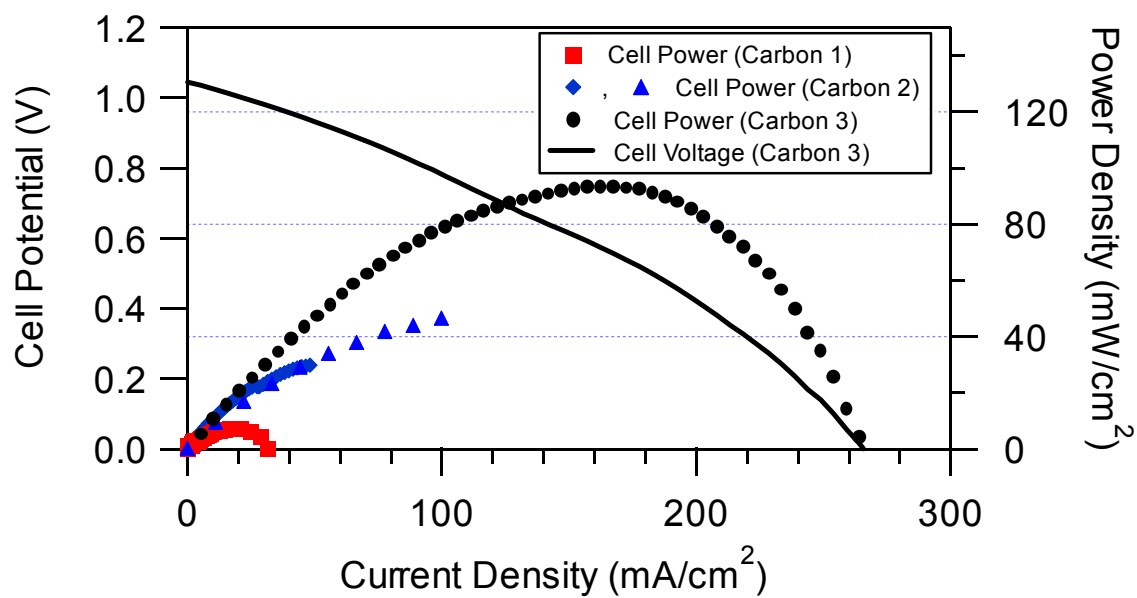


Figure 2. One carbon sample, Carbon 1, shows a peak power near $100 \text{ mW}/\text{cm}^2$.

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